

UDC 662.998:666.3:678:84

SYNTACT HEAT-INSULATING MATERIALS BASED ON CERAMIC-FORMING ORGANOSILICON BINDER

V. Yu. Chukhlanov,¹ É. P. Sysoev,¹ and E. N. Tereshina¹

Translated from *Steklo i Keramika*, No. 5, pp. 25 – 26, May, 2005.

The possibility of using hollow ceramic microspheres and an organosilicon binder with silane and carbosilane links to obtain heat-resistant heat-insulating foam ceramics for construction purposes is demonstrated. The effect of the filler and the gaseous medium on the products of thermal destruction and the properties of the composite is considered. The thermophysical and physicomechanical characteristics of the heat-insulating materials are specified.

Syntact materials (SM) are hollow microspheres bound by a mineral or organic binder, which are promising for use for heat insulation with enhanced physicomechanical and thermophysical characteristics. The study in [1] described compositions based on hollow inorganic microspheres and an organosilicon binder with the siloxane main chain. However, the disadvantage of these heat-insulating materials is their relatively low thermal resistance. Thus, the service temperature for SM with the most heat-resistant polyphenylsiloxane binder is not more than 450°C and after this temperature is exceeded the strength parameters deteriorate catastrophically.

The thermal strength of SM can be increased by using fundamentally new organosilicon binders whose main chain in addition to siloxane links also contains carbosilane links. Until recently it was believed that such compounds can hardly ever be used in the industry [2]. However, it is currently established that the thermal destruction of such polymer binders at temperatures above 600°C produces heat-resistant silicon oxide and silicon carbide while retaining the initial polymer structure (USSR Inventors Certif. No. 1736979). Therefore, if the initial polymer contains a heat-resistant filler, it is possible after heat treatment to obtain a ceramic composite with high strength parameters.

We investigated the problems of the development of heat-insulating SM based on an organosilicon binder for construction purposes with service temperature up to 1200°C. The filler consisted of hollow ceramic microspheres (frequently called cenospheres) produced by flotation treatment of flue gases from thermal power plants operating on solid fuel. The fractional composition of microspheres is given in Fig. 1. The binder is VKL-1 lacquer (TU 6-05-64-101-85)

whose main component is oligooxidehydride-silmethylene-siloxysilane (OHSMS), which besides siloxane links also contains carbosilane and silane links.

The technology of sample preparation consists in mixing the binder with hollow ceramic microspheres up to reaching the “moist sand” consistency, molding the composite at a low pressure, and subsequent thermal treatment. The detailed technology of producing this SM is described in [3].

On the basis of theoretical principles, it can be assumed that intense thermal destruction processes which are accompanied by substantial modifications of the physicomechanical and thermophysical properties of the polymer and its transformation into a ceramic-like state occur at tempera-

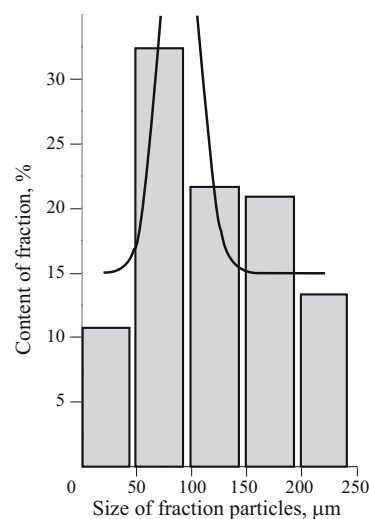


Fig. 1. Fractional composition of analyzed batch of hollow microspheres.

¹ Vladimir State University, Vladimir, Russia.

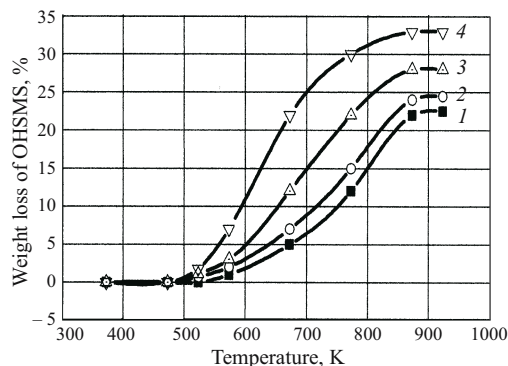


Fig. 2. Weight loss of binder in foam ceramics: 1, 2, 3, and 4) 100, 90, 50, and 10% OHSMS, respectively.

tures above 400–450°C. It can be expected that silanol groups in glass microspheres facilitate the destruction of OHSMS, which is corroborated by thermogravimetric analysis (Fig. 2). The maximum weight loss is not more than 33%.

According to the method in [4], based on the type of the curves it is assumed that the reaction of depolymerization corresponds to the first-order reaction; therefore, the reaction rate constants are calculated based on the following formula:

$$k = \frac{1}{\tau} \ln \frac{c_0}{c_\tau},$$

where τ is the current time; c_0 is the initial concentration; c_τ is the concentration at the moment τ .

As the concentration of OHSMS decreases from 100 to 10%, the depolymerization reaction rate constant grows from 0.00094 to 0.00479 sec^{-1} , i.e., by about 80%. This also corroborates the intensification of destruction processes. Using the reaction rate constants, the activation energy is found from the following formula:

$$E = \frac{\ln(k_1/k_2)RT_1T_2}{T_2 - T_1}.$$

The estimated values of the reaction rate constants (the rate of destruction of the binder) and activation energy are shown in Table 1.

As can be expected, with increasing content of hollow ceramic microspheres in the composite the activation energy sharply decreases. Thus, in the absence of microspheres the thermodestruction activation energy is 23,004.7 J/mole and

TABLE 1

Content of OHSMS, %	k_1 , sec^{-1} ($T_1 = 673$ K)	k_2 , sec^{-1} ($T_2 = 823$ K)	E , J/mole
100	0.0009	0.0016	23,004.7
90	0.0013	0.0018	16,110.6
50	0.0022	0.0029	12,633.6
10	0.0047	0.0053	5024.0

with 90% filler content this energy is reduced to less than one-fourth. The chemical analysis of the products of OHSMS destruction indicates that the main product of the destruction of OHSMS in an oxidizing medium is silicon oxide with a silicon carbide impurity, whereas in an inert medium the products are silicon oxide, carbon, and silicon carbide (Table 2).

Since the products of the thermal destruction of OHSMS are high-melting materials, i.e., silicon oxide and partly silicon carbide, experiments were performed to develop a foam-ceramic heat-insulating SM resistant up to a temperature of 1200°C. Thermal treatment was performed at preset temperatures for 2 h. Up to the preset temperature they were heated at the rate of 5 K/min. Under this regime, no damage was inflicted on the samples by the gaseous products of thermal destruction.

To study the processes occurring in SM at high temperatures and to identify the range of formation of a stable structure, x-ray diffraction analysis was performed on a DRON-2 universal x-ray diffractometer. In deciphering x-ray peaks it was found that the crystalline phase is represented by a perceptible amount of α -quartz, and the rest is the amorphous phase. A further rise in the heat-treatment temperature leads to the processes of thermal destruction accompanied by an abrupt increase in the quantity of the crystalline phase in the SM. Thus, at 1073 K (800°C) a crystalline phase is formed that is mainly represented by mullite and a small quantity of

TABLE 2

Conditions		Mass content of SiO_2 , %, in products of destruction of polymer	
temperature, °C	duration, h	in oxidizing medium (air)	in inert medium (argon)
400	2	28.2	11.2
	4	32.7	13.5
	6	33.6	13.8
	8	35.8	14.1
550	2	59.2	35.1
	4	65.8	38.2
	6	69.9	39.9
	8	72.3	40.1
700	2	80.3	42.5
	4	84.2	45.6
	6	84.5	48.7
	8	85.6	52.5

TABLE 3

Sample*	Apparent density, kg/m^3	Compressive strength, MPa	Thermal conductivity $\text{W/(m} \cdot \text{K)}$
Before heat treatment	380	3.2	0.175
After heat treatment at 900°C	397	6.2	0.182

* Binder content 10 vol.%.

α -quartz. On further temperature rise, the composition of the crystalline phase does not change significantly.

After heat treatment both physicomachanical and thermophysical parameters of heat-insulating foam ceramic materials become modified (Table 3). The strength parameters are doubled and the thermophysical parameters do not change significantly.

Thus, according to the existing classification, syntact materials can be classified as heat-resistant heat-insulating construction materials that can be used in various sectors of industry, in particular in ferrous and nonferrous metallurgy.

REFERENCES

1. V. Yu. Chukhlanov and É. P. Sysoev, "Use of hollow microspheres in organosilicon syntact foam materials," *Steklo Keram.*, No. 2, 11 – 12 (2000).
2. V. Bazhant, V. Khvalovski, and D. Radsuzki, *Silicon* [Russian translation], Khimiya, Moscow (1960).
3. V. Yu. Chukhlanov and A. N. Alekseenko, "Use of syntact foam plastics with organosilicon binders in construction," *Stroitel. Mater.*, No. 6, 26 – 27 (2001).
4. M. T. Bryk, *Destruction of Filled Polymers* [in Russian], Khimiya, Moscow (1989).